

12-17-01

CPA/170 \$
PTO/SB/23 (10-00)**CONTINUED PROSECUTION APPLICATION (CPA)****REQUEST TRANSMITTAL**

Submit an original, and a duplicate for fee processing.

(Only for Continuation or Divisional applications under 37 CFR 1.53(d))

OCT 30 2001

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Address to:

Assistant Commissioner for Patents
Box CPA
Washington, DC 20231

Attorney Docket No. of Prior Application	Mo4532/LeA 31,223
First Named Inventor	Frank Richter et al
Examiner Name	R. Sergeant
Group Art Unit	1711
Express Mail Label No.	ET671450446US

RECEIVED
DEC 19 2001
TC 1700

This is a request for a continuation or divisional application under 37 CFR 1.53(d),
 (continued prosecution application (CPA)) of prior application number 08 / 713,905,
 filed on September 13, 1996, entitled Process for the Production of Ether Isocyanates.

NOTES

FILING QUALIFICATIONS: The prior application identified above must be a nonprovisional application that is either: (1) complete as defined by 37 CFR 1.51(b), or (2) the national stage of an international application in compliance with 35 U.S.C. 371. Effective May 29, 2000, a CPA may only be filed in a utility or a plant application if the prior nonprovisional application was filed before May 29, 2000. A CPA may be filed in a design application regardless of the filing date of the prior application. See "Request for Continued Examination Practice changes to and Provisional Application Practice," Final Rule, 65 Fed. Reg. 50092 (Aug. 16, 2000); Interim Rule, 65 Fed. Reg. 14865 (Mar. 20, 2000), 1233 Off. Gaz. Pat. Office (Apr. 11, 2000).

C-I-P NOT PERMITTED: A continuation-in-part application cannot be filed as a CPA under 37 CFR 1.53(d), but must be filed under 37 CFR 1.53(b).

EXPRESS ABANDONMENT OF PRIOR APPLICATION: The filing of this CPA is a request to expressly abandon the prior application as of the filing date of the request for a CPA. 37 CFR 1.53(b) must be used to file a continuation, divisional, or continuation-in-part of an application that is not to be abandoned.

ACCESS TO PRIOR APPLICATION: The filing of this CPA will be construed to include a waiver of confidentiality by the applicant under 35 U.S.C. 122 to the extent that any member of the public who is entitled under the provisions of 37 CFR 1.14 to access to, copies of, or information concerning, the prior application may be given similar access to, copies of, or similar information concerning, the other application or applications in the file jacket.

35 U.S.C. 120 STATEMENT: In a CPA, no reference to the prior application is needed in the first sentence of the specification and none should be submitted. If a sentence referencing the prior application is submitted, it will not be entered. A request for a CPA is the specific reference required by 35 U.S.C. 120 and to every application assigned the application number identified in such request, 37 CFR 1.78(a).

WARNING: Information on this form may become public. Credit card information should not be included on this form. Provide credit card information and authorization on PTO-2038.

1. Enter the unentered amendment previously filed on _____ under 37 CFR 1.116 in the prior nonprovisional application.
2. A preliminary amendment is enclosed.
3. This application is filed by fewer than all the inventors named in the prior application, 37 CFR 1.53(d)(4).
 - a. **DELETE** the following inventor(s) named in the prior nonprovisional application:
.....
.....
 - b. The inventor(s) to be deleted are set forth on a separate sheet attached hereto.
4. A new power of attorney or authorization of agent (PTO/SB/81) is enclosed.
5. Information Disclosure Statement (IDS) is enclosed: 12/18/2001 STEFFERA 00000141 133848 08713905
01 FC:131 740.00 CH
- a. PTO-1449
- b. Copies of IDS Citations

[Page 1 of 2]

CLAIMS	(1) FOR	(2) NUMBER FILED	(3) NUMBER EXTRA	(4) RATE	(5) CALCULATIONS
	TOTAL CLAIMS (37 CFR 1.16(c) or (i))	6 -20* =		x \$ _____ =	\$ _____
	INDEPENDENT CLAIMS (37 CFR 1.16(b) or (i))	2 -3** =		x \$ _____ =	
	MULTIPLE DEPENDENT CLAIMS (if applicable) (37 CFR 1.16(d))			+ \$ _____ =	
				BASIC FEE (37 CFR 1.16)	740.00
				Total of above Calculations =	
	Reduction by 50% for filing by small entity (Note 37 CFR 1.27).				
	* Reissue claims in excess of 20 and over original patent. ** Reissue independent claims over original patent.			TOTAL =	740.00

6. Small entity status: Applicant claims small entity status. See 37 CFR 1.27.

7. The Commissioner is hereby authorized to credit overpayments or charge the following fees to Deposit Account No. 13 - 3848:

- Fees required under 37 CFR 1.16.
- Fees required under 37 CFR 1.17.
- Fees required under 37 CFR 1.18.

8. A check in the amount of \$_____ is enclosed.

9. Payment by credit card. Form PTO-2038 is attached.

10. Applicant requests suspension of action under 37 CFR 1.103(b) for a period of _____ months (not to exceed 3 months) and the fee under 37 CFR 1.17(i) is enclosed.

11. New Attorney Docket Number, if desired
(Prior application Attorney Docket Number will carryover to this CPA unless a new Attorney Docket Number has been provided herein.)

12. a. Receipt For Facsimile Transmitted CPA (PTO/SB/29A)
b. Return Receipt Postcard (Should be specifically itemized. See MPEP 503)

13. Other:

NOTE: *The prior application's correspondence address will carryover to this CPA
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14. NEW CORRESPONDENCE ADDRESS

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15. SIGNATURE OF APPLICANT, ATTORNEY, OR AGENT REQUIRED

Name (Print / Type) Lyndanne M. Whalen

Signature Lyndanne M. Whalen

Registration No. (Attorney/Agent) 29,457

Date October 30, 2001

THE COMMISSIONER OF PATENTS AND TRADEMARKS

Transmitted herewith for filing is the patent application of 1) Frank Richter; 2) Josef Pedain
3) Klaus Neichtkamp; 4) Andreas Flink

FOR THE PRODUCTION OF ETHER ISOCYANATES

ENCLOSED ARE THE FOLLOWING PAPERS:

- Specification WITH attached Declaration
- Specification WITHOUT attached Declaration
- _____ sheets of FORMAL/INFORMAL drawings containing Figures (s) _____
- Preliminary Amendment

Enclosed herewith are copies of documents identified in the present application as related to the Background of the Invention for the Examiner's consideration under 37 CFR 1.104(a). The attached Form PTO 1449 identifies the submitted art.

An Assignment of the Invention to BAUER AKTIENGESELLSCHAFT Please charge the fee for recording the Assignment, \$40.00, to Deposit Account Number 13-3848.

Any notice that is to be furnished to the above organization after grant of the patent relating to maintenance fees or for any other reason should be addressed to:

Bayer Corporation Telephone: (412) 777-2349
Patent Department
100 Bayer Road
Pittsburgh, Pennsylvania 15205-9741

Priority is hereby claimed based upon the following application(s), a certified copy of each is being submitted herewith:
Federal Republic of Germany Application Number(s) 195 35 506.7
filed in Germany September 25, 1995

THE TOTAL AMOUNT DUE FOR THE FILING FEE IN THIS CASE IS:

CLAIMS AS FILED				
FOR	NUMBER FILED	NUMBER EXTRA	RATE	Fee
TOTAL CLAIMS	4 - 20		X \$.22 =	\$
INDEPENDENT CLAIMS	2 - 3		X \$.78 =	
MULTIPLE DEPENDENCY FEE			+ \$ 250 =	
BASIC FEE				\$ 750.00

TOTAL FILING FEE \$ 750.00
Plus fee for recordal of Assignment - \$ 40.00

- Please charge Deposit Account Number 13-3848 in the amount of \$ 790.00. Three copies of this sheet are enclosed.
- The Commissioner is authorized to charge any fee due under 37 CFR 1.16 and 1.17 during the entire pendency of this application or credit to our Deposit Account Number 13-3848.
- This is a general authorization EXCEPT that this is NOT AN AUTHORIZATION FOR PREPAYMENT OF AN ISSUE FEE or any other fee under 37 CFR 1.18.

Date September 13, 1996

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Date of Report September 13, 1996

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Donna J. Veatch

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John Vela
(Signature of person mailing paper or free)



Mo4532
LeA 31,223 -US

PROCESS FOR THE PRODUCTION OF ETHER ISOCYANATES

BACKGROUND OF THE INVENTION

5 The present invention relates to a process for the production of ether (poly)isocyanates by phosgenation of ether amines in the vapor phase.

Isocyanates containing ether groups or ether isocyanates are well known. (See, for example, *Annalen der Chemie*, 562 (1949), 83 ff).

10 However, these known ether-containing isocyanates are generally obtainable only in poor yield and low purity.

In the reactions of ether amines, chlorinated products are frequently obtained by splitting the ether (U.S. Patent 3,267,122). Thus, for example, $\text{H}_2\text{N}(\text{CH}_2)_3-\text{O}-(\text{CH}_2)_4-\text{O}-(\text{CH}_2)_3\text{NH}_2$ is split into $\text{OCN}(\text{CH}_2)_3\text{Cl}$ 15 during the reaction with COCl_2 (reference: *Angew. Chem.*, A 59 (1949), 271).

Only aliphatic ether amines having HCl salts which are soluble in chlorinated hydrocarbons, for example, $\text{C}_4\text{H}_9-\text{O}-(\text{CH}_2)_3\text{NH}_2$, can be converted to ether isocyanates at temperatures below 80°C (*Annalen der Chemie*, 562 (1949), 105). The yield of this isocyanate is, however, only 86% of the theoretical yield. In such a process, it is necessary to convert the amine into the amine hydrochloride prior to the phosgenation. About 20 80% methoxypropylamine hydrochloride splits at 140 to 150°C in 1-chloronaphthalene with phosgene to form 3-chloropropyl isocyanate 25 (*Annalen der Chemie*, 562 (1949), 104).

Certain ether isocyanates can be obtained in yields of up to about 30 80% by simple base phosgenation. (See, for example, DE-A 1,154,092.) However, the products of such processes have very high residual chlorine contents (0.1%). Such a high chlorine content in the diisocyanate frequently makes it difficult to use those products. For example, such chlorine-containing diisocyanates are not useful for the preparation of

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Donna J. Vearch

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Donna J. Vearch

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non-discoloring raw materials for coatings. The corresponding hydrochlorides of the amines or carbamates must be used in such processes. The handling of heterogeneous reaction mixtures of this type is, however, very difficult and is an obstacle to the smooth, economical

5 production of the isocyanates.

(Poly)isocyanates containing ether groups are also obtainable by nucleophilic substitution of organic halides by metal cyanates. (See, for example, JP 50 036 424; Arch. d. Pharm., 302 (1969), 617; and DE-A 2,031,291) The accumulation of salts, the generally low conversion rates

10 and the environmental problems encountered in these processes are obstacles to the industrial exploitation of this approach.

The reaction of ether (poly)amines with low-molecular weight alkyl isocyanates, subsequent thermal decomposition of the ureas formed and separation of alkylamine has also been proposed as a method for the

15 preparation of ether (poly)isocyanates ("isocyanate interchange"; see, for example, DE-A 3,232,917). However, this method has several disadvantages. First, a by-product which must be disposed of is obtained. Second, considerable quantities of urea remain in the product, particularly when the ether (poly)isocyanates cannot be worked up by

20 subsequent purification processes. Another disadvantage of this process is that the "isocyanate interchange" is a typical equilibrium reaction and is therefore difficult to carry out quantitatively.

Isocyanates containing ether groups can also be prepared by Curtius rearrangement of the corresponding carboxylic acid azides

25 (J. Prakt. Chem., 335 (1993), 294 and the references cited therein), but only on a laboratory scale.

As is explained in DE-A 1,165,580, e.g., polyisocyanates containing ether groups are of great interest for use in paints and coatings.

Splitting the ether group in ether amines occurs more readily and

30 completely at elevated temperatures. For example, phosgenation of 3-

methoxypropylamine in toluene below 110°C produces a mixture of 3-methoxypropyl isocyanate and 3-chloropropyl isocyanate. At elevated temperature (e.g., 140 to 150°C) in chloronaphthalene as solvent, however, substantially only 3-chloropropyl isocyanate is formed (Annalen der Chemie, 562 (1949), 83).

5 DE-A 1,793,329 discloses a cold phase-hot phase phosgenation in solution for the preparation of ether(poly)isocyanates. It is alleged that very little, if any, splitting of the ether occurs. However, the yields of isocyanate are only 60 to 75% of the theoretical yield. The chlorine 10 content of the products, at 400 to 2000 ppm, is far too high for many applications, particularly for paint and coatings applications.

SUMMARY OF THE INVENTION

It is an object of the present invention to provide a process for the production of ether (poly)isocyanates.

15 It is also an object of the present invention to provide a simple process for the production of isocyanates containing ether groups.

It is another object of the present invention to provide a process for the production of high quality isocyanates containing ether groups.

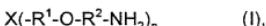
20 It is a further object of the present invention to provide a process for the production of high quality ether isocyanates in high yields without significant product loss through, for example, splitting of the ether groups.

These and other objects which will be apparent to those skilled in the art are accomplished by converting mono- and polyamines containing ether groups to the corresponding isocyanates in very good yields and in 25 high purity, without splitting the ether group. This conversion is achieved by reacting an ether-containing amine in the vapor phase with phosgene in the vapor phase under applied pressure at a temperature in the range from 50 to 800°C, preferably from 100 to 550°C (depending on the boiling point of the amine), optionally in the presence of an inert carrier 30 gas.

DETAILED DESCRIPTION OF THE INVENTION

The present invention relates to a method for preparing ether (poly)isocyanates from ether (poly)amines in which ether (poly)amines are reacted with at least the stoichiometric quantity, based on the NH₂ group(s), of phosgene or corresponding quantities of a material which generates phosgene, in the vapor phase close to or above the boiling point of the starting (poly)amine which boiling point is in the temperature range of from 50 to 800°C, preferably from 100 to 550°C, under applied pressure.

10 In the present invention, the ether amines which may be used include compounds represented by Formula (I)



in which

X represents H, NH₂ or C(R³)_{4-n},

15 R¹, R² and R³ each represent the same or a different, optionally branched, optionally substituted (e.g., with Cl, Br), optionally heteroatom-containing (e.g., N, O, S) C₁-C₁₀alkyl, C₃-C₂₄ cycloalkyl, C₇-C₂₄ aralkyl, or C₆-C₂₄ aryl radical, and R¹ may also represent a direct bond between X and the ether oxygen atom bonded to R²,

20 and

n represents 1, 2 or 3.

The process of the present invention may be carried out using known techniques. Suitable techniques are disclosed in EP-A 0,570,799 and DE-A 4,412,327. In these disclosed processes, the co-reactants are introduced into suitable reactors maintained at a temperature close to or above the boiling point of the starting amine or mixture of amines. The co-reactants are then mixed and reacted with one another. The temperature, depending upon the pressure, is generally between 50 and

800°C, preferably between 100 and 550°C. The process is generally carried out within a pressure range of from 10 mbar to 5 bar, preferably from 200 mbar to 3 bar.

Introduction of the reaction components during the vapor phase

5 phosgenation may optionally take place in the presence of inert additives such as carrier gases. The carrier gases used may be nitrogen, argon or other inert gases and vapors of commercially available solvents such as chlorobenzene, dichlorobenzenes, xylenes, chloronaphthalenes and decahydronaphthalene.

10 The phosgene used in the phosgenation reaction is used in a stoichiometric amount or in stoichiometric excess, determined on the basis of the number of primary amino groups in the amine starting material. A quantity of phosgene amounting to from 100 to 300% of the theoretical quantity, preferably from 100 to 200% of the theoretical

15 quantity, is generally sufficient.

After the reaction with phosgene, the ether isocyanates are recovered by cooling the gas stream to a temperature above the decomposition temperature of the corresponding intermediate carbamic acid chlorides. The ether isocyanate may then be isolated in pure form
20 by known processes such as distillation, crystallization, extraction or film distillation, or recovered as raw product (solution).

The amine starting materials which are converted into the corresponding isocyanates by the process of the present invention may be obtained by a number of known processes. One suitable known
25 process is alkoxylation of water or of other, optionally polyfunctional, OH-functional compounds such as alcohols, phenols and/or carboxylic acids and subsequent amination (for example, FR-A 1 361 810). Another suitable process for producing the amine starting material is polymerization of tetrahydrofuran and, optionally after further reaction
30 with alkylene oxide, subsequent treatment as described in FR-A-1 361

810. Suitable amine starting materials may also be produced by cyanoethylation of water and subsequent hydrogenation to form bis(3-aminopropyl)ether (DRP 731 708) or by cyanoethylation of other, optionally polyfunctional, OH-functional compounds (particularly diols and
5 triols) and subsequent hydrogenation.

The usefulness of mono- and polyamines containing ether groups in the phosgenation of the present invention is determined essentially by the vapor pressure of the amine at the applied pressure. In the case of particularly high-boiling compounds, it may be advantageous to introduce
10 the amine into the phosgenation reaction as an azeotrope with other substances, or to use a carrier gas for the introduction of the amine component into the reaction chamber.

Typical examples of suitable (poly)amines of Formula (I) which may be used alone or as mixtures include: alkyl aminoalkyl ethers such
15 as aminomethyl methyl ether, aminomethyl ethyl ether, aminomethyl propyl ether (as well as isomers), 1-aminoethyl methyl ether, 2-aminoethyl methyl ether, and aminopropyl methyl ether (as well as isomers); diamino-oxoalkanes such as 1,1'-bis(aminomethyl) ether, 1,1'-bis(aminoethyl) ether, 1,2'-bis(aminoethyl) ether, 2,2'-bis(aminoethyl) ether and
20 technical mixtures of the three latter diamines, bis(aminopropyl) ether (all isomers, optionally as a mixture), diamino(poly)oxoalkanes such as 1,8-diamino-1,5,8-trimethyl-3,6-dioxaoctane, 1,11-diamino-1,5,8,11-tetramethylundecane and all isomers of the two latter compounds having vicinal O-N bonding in pure form or as a mixture (for example, as
25 commercial Jeffamine D 230), 1,8-diamino-3,6-dioxaoctane (for example, as commercial Jeffamine EDR 148), 1,10-diamino-4,7-dioxadecane, 1,12-diamino-4,9-dioxadodecane, 1,14-diamino-3,10-dioxatetradecane, and 1,13-diamino-4,7,10-trioxatridecane; triamino(poly)oxoalkanes such

as 1,7-diamino-2,6-dioxa-4-aminomethoxyheptane, 1-amino-2-oxa-3,3-bis(aminomethoxy)hexane, 1,9-diamino-3,7-dioxa-5-(1-amino-2-ethoxy)-nonane, 1-amino-3-oxa-4,4'-bis(1-amino-2-ethoxy)heptane, 1,11-diamino-4,8-dioxa-6-(1-amino-5-oxobutyl)undecane, 1-amino-4-oxa-5,5-bis(1-

5) amino-5-oxobutyl)octane and mixtures of the above-mentioned monoamines, diamines and triamines.

The mixture of isomers composed of 2-(2-isocyanatopropoxy)-1-propyl isocyanate, 1,1'-oxydi-2-propyl isocyanate and 2,2'-oxydi-1-propyl isocyanate ("dipropylene glycol diisocyanate", mixture of isomers)

10 prepared by the process of the present invention is new.

The ether isocyanates prepared by the process of the present invention are valuable raw materials for the production of polyurethanes (optionally foamed), adhesives, coating materials, emulsifiers, thickeners, oligomeric isocyanate modification products (e.g., polyisocyanates
15 containing uretdione, isocyanurate, carbodiimide, biuret, urethane and allophanate groups), and auxiliary substances which are used, for example, for imparting wet strength to paper and other cellulose products. These ether isocyanates are useful as raw materials for the production and/or formulation of active substances and pharmaceuticals
20 (DE-A 3,232,917).

The invention is further illustrated but is not intended to be limited by the following examples in which all parts and percentages are by weight, unless otherwise specified.

EXAMPLES

25 Example 1

2-(2-isocyanatopropoxy)-1-propyl isocyanate, 1,1'-oxydi-2-propyl isocyanate and 2,2'-oxydi-1-propyl isocyanate ("dipropylene glycol diisocyanate", mixture of isomers) were prepared by the procedure described below.

The apparatus in which the reaction was conducted included a mixer tube heated to 400°C which was 2.5 mm in diameter and 17.5 mm in length having a condensation stage arranged in tandem and a connected COCl₂ adsorption tower filled with activated carbon. COCl₂,

- 5 which had been heated to 420°C at 950 mbar in a heat exchanger connected in front, flowed continuously at a rate of 2.5 mol/h through a nozzle projecting into the mixer tube. Simultaneously, a mixture of amines heated to 320°C, obtained by catalytic amination under pressure of technical dipropylene glycol (approx. 50% 2-(2-hydroxypropoxy)-1-
- 10 propanol, approx. 40% 1,1'-oxydi-2-propanol and approx. 10% 2,2'-oxydi-1-propanol) having a boiling range of 72 to 78°C at a pressure of 7.5 mbar, was introduced at a feed rate of 1 mol/h, together with dry nitrogen at a rate of 0.1 mol/h as diluent, into the reaction chamber via the annular passage between the nozzle and the mixer tube. A pressure of
- 15 approx. 350 mbar was maintained in the mixer tube by applying a reduced pressure at the end of the condensation stage. That is, the reaction mixture leaving the reaction chamber was passed in a condensation stage through 1,2-dichlorobenzene, which was maintained at 150 to 160°C. Here the selective condensation of the diisocyanates
- 20 formed took place. In the adsorption tower, COCl₂ was separated from the gas mixture passing through the scrubbing stage and containing nitrogen, HCl and excess COCl₂. The mixture of diisocyanates was recovered in a pure state by distillation ($K_p = 95^\circ\text{C}/0.05 \text{ mbar}$, $n_D = 1.4393/20^\circ\text{C}$) and descended as a colorless liquid having an NCO content, titrated in accordance with DIN 53 185, of 45.4% (theoretical: 45.6%). The yield of the pure, distilled mixture of diisocyanates was 98.2% of the theoretical yield, based on the mixture of diamines used, with a purity of 99.7% as determined by gas chromatography and a content of hydrolyzable chlorine of 43 ppm.
- 25
- 30 Example 2

1,8-diisocyanato-3,6-dioxaoctane was prepared in accordance with the procedure described below.

2.5 kg (16.87 mol) of 1,8-diamino-3,6-dioxaoctane which is commercially available from Aldrich (also known as Jeffamine EDR 48) 5 was converted into 1,8-diisocyanato-3,6-dioxaoctane and isolated in the manner specified in Example 1.

Yield: 3360 g = 99.5% of the theoretical yield, purity (GC): 99.8%, NCO content in accordance with DIN 53 185: 42.0% (theoretical: 42.0%), Hydrolyzable chlorine content: 48 ppm. Kp: 95°C/0.5 mbar.

10 Example 3

1,12-diisocyanato-4,9-dioxadodecane was prepared from 2.5 kg (12.24 mol) of 1,12-diamino-4,9-dioxadodecane (commercially available from Aldrich) and isolated in the manner specified in Example 1.

Yield: 3056 g = 97.4% of the theoretical yield, purity (GC): 99.5%, 15 NCO content in accordance with DIN 53 185: 32.9% (theoretical: 33.0%), Hydrolyzable chlorine content: 34 ppm. Kp: 83°C/0.2 mbar.

Example 4

1,3-bis(3-isocyanatopropoxy)-2,2-dimethyl propane was prepared from 2.5 kg (11.45 mol) of 1,3-bis(3-aminoproxy)-2,2-dimethyl propane 20 (commercially available from Aldrich) and isolated in the manner specified in Example 1.

Yield: 3067 g = 99.1% of the theoretical yield, purity (GC): 99.8%, NCO content in accordance with DIN 53 185: 31.0% (theoretical: 31.1%), Hydrolyzable chlorine content: 24 ppm. Kp: 108°C/0.1 mbar.

25 Example 5

3-methoxypropyl isocyanate was prepared from 1,000 g (11.2 mol) of 3-methoxypropylamine (commercially available from Aldrich) was converted into the isocyanate and isolated in the manner specified in Example 1.

Yield: 1250 g = 96.8% of the theoretical yield, purity (GC): 99.1%, NCO content in accordance with DIN 53 185: 36.5% (theoretical: 36.50%), Hydrolyzable chlorine content: 44 ppm. Kp: 55°C/20 mbar.

The identity of all the compounds produced in these Examples
5 was deduced from IR, ¹H-NMR, ¹³C-NMR and mass spectroscopic analyses and from the results of elemental analysis.

Comparative examples (liquid phosgenation)

10 440 g of monochlorobenzene were mixed at 5°C with 330 g of phosgene in a four-necked mixing flask equipped with reflux condenser, internal thermometer, dropping funnel and inlet tube. Then a solution of 71.5 g of the mixture of diamines specified in Example 1 in 900 g of monochlorobenzene was added dropwise over a period of 90 min.

15 The reaction mixture was then slowly heated, with stirring, to an internal temperature of 90°C with simultaneous introduction of phosgene (approx. 1 mol/h) and maintained at this temperature for several hours. It was not possible to achieve a complete elucidation of the reaction mixture. After blowing off of the excess phosgene with nitrogen, filtration and working up by distillation, 19.5 g (19.6% of the theoretical yield) of a slightly colored liquid was obtained, having a boiling range of 80 to 20 85°C/0.07 mbar and an NCO content in accordance with DIN 53 185 of 45.2%.

25 Neither varying the solvent (1,2-dichlorobenzene (34% theoretical yield) and toluene (22.3% theoretical yield)) nor converting the mixture of diamines into the dihydrochloride and bis(carbamate) (15.3% and 27.6% yields, respectively) increased the yield of diisocyanate substantially. The residual chlorine content of the product was in no case below 0.1%.

Further examples of phosgenations of ether diamines in the liquid phase are described, for example, in Annalen der Chemie, 562 (1949), 6 ff; DE-A 1,154,092; JP 4,027,365; FR 1,578,622.

Although the invention has been described in detail in the foregoing for the purpose of illustration, it is to be understood that such detail is solely for that purpose and that variations can be made therein by those skilled in the art without departing from the spirit and scope of the invention except as it may be limited by the claims.

WHAT IS CLAIMED IS:

Su D B

1. A process for the production of an ether (poly)isocyanate from an ether (poly)amine comprising reacting
 - a) an ether (poly)amine
- 5 with at least a stoichiometric amount (based on the number of primary amine groups present in a)) of
 - b) phosgene or a compound which generates phosgene under the reaction conditions
- in the vapor phase at a temperature of from about 50 to about 800°C
- 10 which temperature is close to or above the boiling point of a) under applied pressure.
2. The process of Claim 1 in which ether (poly)amine a) is represented by the formula

$$X-(R^1-O-R^2-NH_2)_n \quad (I)$$

15 in which

X represents H, NH₂ or C(R³)_{4-n}.

R¹, R² and R³ each represents an optionally branched, an optionally substituted, or an optionally heteroatom-containing C₁-C₁₀ alkyl, C₃-C₂₄ cycloalkyl, C₇-C₂₄ aralkyl, or a C₆-C₂₄ aryl radical, provided that R¹ may also represent a direct bond of X to the ether oxygen atom bonded to R²,

20 and

25 n represents 1, 2 or 3.

3. The ether (poly)isocyanates selected from 2-(2)isocyanato-propoxy)-1-propyl isocyanate, 1,1'-oxydi-2-propyl isocyanate, 2,2'-oxydi-1-propyl isocyanate and mixtures thereof.

4. A process for the production of a polyurethane comprising reacting the ether (poly)isocyanate produced in accordance with Claim 1 with an isocyanate-reactive material.

As a below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below next to my name. I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled

PROCESS FOR THE PRODUCTION OF ETHER ISOCYANATES

the specification of which is attached hereto,

or was filed on _____ as

Application Serial No. _____

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims.

I acknowledge the duty to disclose to the Office all information known to me to be material to patentability as defined in Title 37, Code of Federal Regulations, §1.56.

I hereby claim foreign priority benefits under Title 35, United States Code, §119 of any foreign application(s) for patent or inventor's certificate listed below and have also identified below any foreign application for patent or inventor's certificate having a filing date before that of the application on which priority is claimed:

Prior Foreign Application(s), the priority(ies) of which is/are to be claimed:

19535506.7 Germany September 25, 1995
(Number) (Country) (Month/Day/Year Filed)

I hereby claim the benefit under Title 35, United States Code, §120 of any United States application(s) listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States application in the manner provided by the first paragraph of Title 35, United States Code, §112, I acknowledge the duty to disclose to the Office all information known to me to be material to patentability as defined in Title 37, Code of Federal Regulations, §1.56 which became available between the filing date of the prior application and the national or PCT international filing date of this application:

(Application Serial No.)	(Filing Date)	(Status) (patented, pending, abandoned)
(Application Serial No.)	(Filing Date)	(Status) (patented, pending, abandoned)

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

POWER OF ATTORNEY: As a named inventor, I hereby appoint the following attorney(s) and/or agent(s) to prosecute this application and transact all business in the Patent and Trademark Office connected therewith:

JOSEPH C. GIL, Patent Office Registration Number 26,602

ARON PREIS, Patent Office Registration Number 29,426

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